

UNCLASSIFIED PRELIMINARY DATA

Report No. IITRI-C6018-11
(Quarterly Report)

INVESTIGATION OF LIGHT SCATTERING
IN HIGHLY REFLECTING PIGMENTED COATINGS

National Aeronautics
and Space Administration

IIT RESEARCH INSTITUTE

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April 1, 1964 to August 1, 1964

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Prepared by

G. A. Zerlaut, V. Raziunas, and S. Katz

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IIT RESEARCH INSTITUTE
Technolgy Center
Chicago 16, Illinois

to

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FOREWORD

This is Report No. IITRI-C6018-11 (Quarterly Report) of Project C6018, Contract No. NASr-65(07), entitled "Investigation of Light Scattering in Highly Reflecting Pigmented Coatings." This report covers the period from April 1, 1964 to August 1, 1964. Previous Quarterly Reports IITRI-C6018-3, IITRI-C6018-6 and IITRI-C6018-8 were prepared on October 11, 1964, January 29, 1964, and May 5, 1964, respectively.

Major contributors to the program include Gene A. Zerlaut (Project Leader), Dr. S. Katz and Dr. B. Kaye (theoretical analyses), V. Razinuas (principal investigator), and Mrs. J. Allen (silver halide preparations).

Data are recorded in Logbooks C14085 and C13906.

Respectfully submitted,



Gene A. Zerlaut
Research Chemist-Group Leader
Polymer Research Section

APPROVED BY:



Theodore H. Meltzer,
Manager
Polymer Research Section

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INVESTIGATION OF LIGHT SCATTERING
IN HIGHLY REFLECTING PIGMENTED COATINGS

ABSTRACT

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The optical properties of concentrated films of suspensions of silver bromide particles of two narrow size distributions was established. For the concentrated suspensions in thin films, it has been shown that if the films are monolayers, the interactions due to proximity of scatterers are minimal. However, multiple scattering effects are found if the particles are packed behind each other with respect to the incident beam.

In dilute systems the particles of two sizes act as independent scatterers. It was thus shown that

$$\frac{D_1 + D_2}{2} = D_{1+2}$$

where D_1 , D_2 and D_{1+2} are the optical densities of suspensions of size 1, size 2 and 50/50 mixture of sizes 1 and 2.

On the basis of experimental data, a theoretical optimum paint coating was constructed in order to attain minimum interactions between the scatterers. The particles should be suspended in exceedingly thin layers containing particles of uniform size. The distribution of layers with respect to the incident beam should be such that each layer contains increasingly larger particles. The top layers, containing the smallest particles, will interact only with shortest wavelengths, allowing longer wavelengths to pass undisturbed in both directions.

Author

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I. INTRODUCTION

The principal objective of this program is the application of light-scattering theory to particle arrays in an attempt to explain the scattering behavior of polydisperse pigmented coatings, especially highly reflecting paint systems. Of particular concern therefore are the definitions of the light-scattering parameters which operate in pigmented coatings designed for the maximum reflection of solar radiation -- a problem of some magnitude since the sun's energy encompasses the rather large wavelength interval from about 2500 to 40,000 Å.

Previous quarterly reports have discussed the preparation on mono-sized silver bromide suspensions and have dealt principally with the optical properties of both mono- and bimodal dispersions of silver bromide particles in liquid and gelatin suspensions.

This report continues the discussion of light-scattering theory. Experimental studies during the period covered by this report have involved the establishment of the optical properties of concentrated films of silver bromide particles of two narrow size distributions. These include the effects of solid angle and path length on transmittance measurements and the evaluation of the transmittance of bimodal suspensions of silver bromide particles.

II. LIGHT-SCATTERING THEORY AND MULTIPLE-SCATTERING SYSTEMS

A. The Application of Light-Scattering Theory to Reflective Coatings

In the following discussions, the application of single-particle scattering theory to multiple particle arrays is considered. The previous discussion of particles with very large refractive indices is extended to include the case of larger particles and multiple particle systems.

B. Multiple Particle Scattering

In previous reports, we have discussed light-scattering theory with reference to single particles. Single-particle theory can be used to describe the light-scattering characteristics of a colloidal or aerosol suspension in which the particles are very widely dispersed. In an application such as the present one where we wish to apply scattering theory to the design of a coating material, it becomes necessary to attempt to set a lower limit to the separation distance which still permits application of scattering theory.

The theoretical problem of computing the scattering of light by a multiple array of particles in terms of the interaction of an electromagnetic wave on a multiple discontinuity does not appear to have been analyzed rigorously. Van de Hulst noted the problem in 1946,¹ and commented then on its complexity.

¹Van de Hulst, H. C., "Optics of Spherical Particles," N. V. Drukkerij, J. J. Duwaer and Sons, Amsterdam, 1946.

A number of references in the literature give an empirical or an estimated partial solution of the problem. Sinclair² suggested a spacing of 10 or preferably 100 times the radius of the particle for independent scattering. However, Berry³ studied the scattering of light by cubic silver bromide crystals embedded in gelatin and reported that the approximation to single independent scattering appears to be quite satisfactory when the separation of the grain centers is more than about twice the grain size.

In previous reports, we defined the term S as the effective scattering area of a single particle. K , the scattering co-efficient in the scattering per unit cross-sectional area of the particle is therefore equal to $S/\pi r^2$. Figure 1 of Report No. 3⁴ is a plot of K against the parameter $\alpha = 2\pi r/\lambda$ for several real refractive indices.

In the absence of a more exact procedure, it is proposed that for particles whose scattering coefficient K is greater than unity, the scattering cross-section, S , governs the limit of particle spacing for effective application of light-scattering

²Sinclair, D., "Handbook on Aerosols," Chapter VII, Atomic Energy Commission, Washington, D.C., 1950.

³Berry, C. R., J. of the Optical Society of America, Vol. 52, 888, 1962.

⁴Report No. IITRI-C6018-3 (Quarterly)

theory. The electromagnetic fields which define the scattering cross-sections will interfere if the particles are spaced more closely. Also, in the case of very small particles where K is less than unity, the scattering cross-section is smaller than the geometric cross-section and the physical dimensions of the particle will dictate the limits of particle spacing.

C. Scattering by Particles With High Refractive Index

The scattering of light by small particles with very large refractive indices, i.e., totally reflecting spheres, was discussed previously.⁵ In this report, the problem in the case of larger particles is examined, and some application to coatings are considered.

Very small particles with infinite refractive indexes are not true Rayleigh scatterers. The radial distribution of the scattered intensity is never symmetrical, but instead the back-scattered intensity at 180° from the forward direction is nine times the intensity of the forward-scattered light. This is a desirable condition for back-scattering of course; unfortunately very small totally reflecting particles have small effective cross-sections and are not much more efficient light scatterers than are Rayleigh scatterers of comparable size. With increasing particle size, the total scattering efficiency increases to

⁵Report No. IITRI-C6018-6

a maximum slightly in excess of two as α ($2\pi r/\lambda$) approaches unity and remains near that value as α increases (Figure 1).

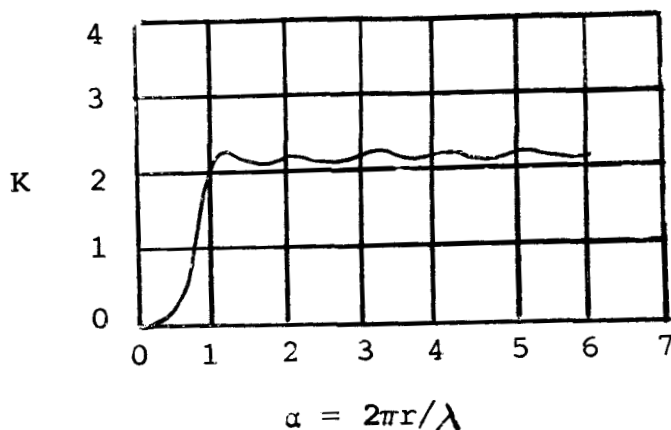


Figure 1
Perfect Reflectors ($M \rightarrow \infty$)

Radial scattering data for larger totally reflecting particles have been given by van de Hulst. It is noted that the strong attenuation of the back-scattered signal which characterized transparent particles is not present here and as might be expected, a large component of energy is present in the back-scattered radiation.¹

An array of totally reflecting particles should therefore produce a substantial attenuation with a relatively small concentration in comparison with transparent particles.

The optimum particle for the present application would, therefore appear to be a particle with an infinite refractive index. In the next report, experimental approximations to such materials will be discussed, and an attempt will be made to construct a theoretical pigment film designed for the selective transmission or rejection of different spectral regions.

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III. EXPERIMENTAL STUDIES

A. Introduction

In the last report⁶ we established some quantitative relationships between the particle concentration, the particle size, and the optical properties of dilute suspensions and concentrated monodisperse films containing silver bromide particles as scatterers. In dilute suspensions of a mixture of two particle sizes, the particles behaved as independent scatterers; the attenuation of light due to each particle size was additive. The scattering properties of the mixture consisted of the sum of the properties of each component of the mixture. For the concentrated suspensions in thin films, it was shown that if the films are monolayers, the interactions due to proximity of scatterers are minimal. On the other hand, multiple scattering effects were found if the particles were packed behind each other with respect to the incident beam.

The immediate objective of experimental studies during this research period was the establishment of the optical properties of concentrated films of suspensions of silver bromide particles of two narrow size distributions. Such a system is the first approximation to a real polydisperse coating. A

⁶Zerlaut, G. A., "Investigation of Light Scattering in Highly Reflecting Pigmented Coatings," Quarterly Report No. IITRI-C6018-8, IIT Research Institute, Chicago, Ill., May, 1964.

successive addition of particles with a narrow size distribution should lead to an actual, highly controlled, polydisperse system in which the physical and optical parameters can be fully defined. The experimental apparatus, and silver bromide particles, and measurement techniques used in this study were described in our last report.⁷ The optical densities are given to the base of natural log.

B. Effects of Solid Angle and Path Length on Transmittance Measurements

In an ideal scattering system in which the conditions of the single-particle theory are met (no absorption and no reflection at the interfaces between the suspension medium and air and also scattering particles), agreement between theory and observation will depend on the solid angle of light flux reaching the detector after interaction with the scatterer. A larger solid angle at the detector will allow a larger number of the photons, which should be lost according to the transmission equation to reach the detector. Thus, as the scattering system is brought closer to the detector, the deviations from the theory should increase. In contrast, if the light losses were due to an absorptive mechanism (Beer's or Lambert's law), the position of the cell would have no effect whatsoever, since the light is

⁷ IIT Research Institute, loc. cit.

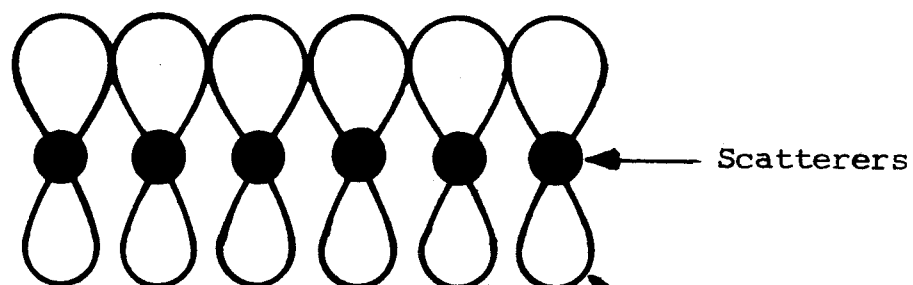
inherently lost and converted to thermal and molecular energy.

The effect of positioning the cells with respect to the detector was qualitatively examined. As predicted by the theory, the amplitude of scattering extrema decreased as the cell was brought closer to the detector.

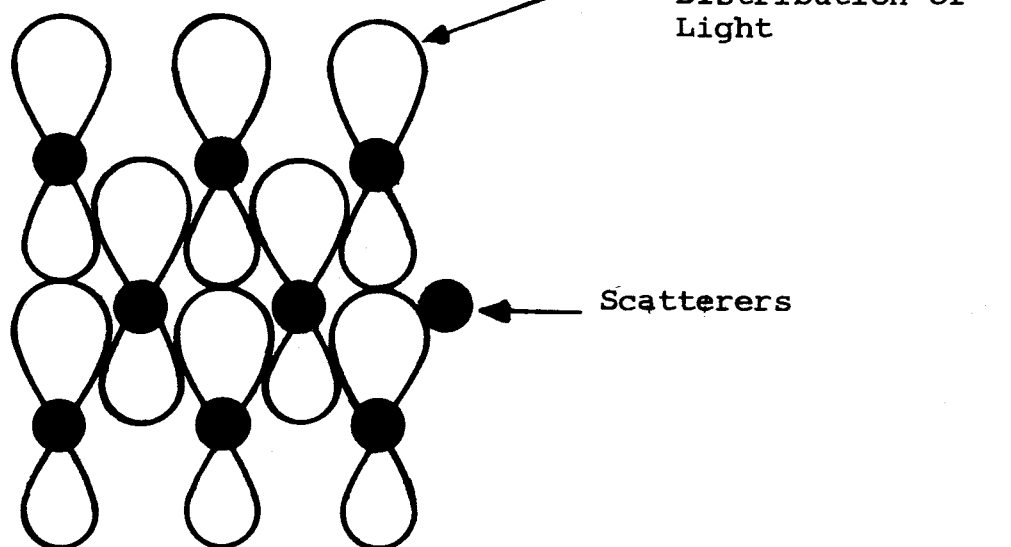
It was previously observed⁸ that packing of particles behind each other with respect to the incident beam tends to give larger deviations from theory than packing even very closely in monolayers. This was explained on the basis of radial distribution of scattered light. A minimum in the radial intensity functions usually occurs at 90° from the incident beam, thus multiple interactions between particles packed in monolayers would be minimal. This observation is schematically illustrated in Figure 2. The general conclusion from the above observation is that increasing optical path length under given conditions would give a larger number of particles projected behind each other and would result in more multiple interactions. Thus, in any real scattering system of appreciable concentration, an increasing optical path length would give increasing deviations from the theory. The results of this study are summarized in Table 1, indicating an appreciable decrease in the ratio of scattering extrema with increase in path length.

⁸IIT Research Institute, loc. cit., pp 29, 31.

a. Concentrated Monolayer



b. Several Layers



↑
Direction of the
Incident Beam

Figure 2

SCHEMATIC DIAGRAM OF THE OVERLAP OF RADIAL INTENSITY FUNCTIONS

Table 1

THE AMPLITUDE OF THE SCATTERING EXTREMA
AS A FUNCTION OF PATH LENGTH

<u>Batch No.</u>	<u>Particle Radius, r</u>	<u>Optical Path Length, cm</u>	<u>Ratio $\frac{1st\ Max.}{1st\ Min.}$</u>
25	0.39	2	1.85
		5	1.55
26	0.32	2	1.61
		5	1.45
28	0.44	2	1.82
		5	1.49
30	0.20	2	1.87
		5	1.60
31	0.33	2	2.09
		5	1.73
Theoretical	-	-	2.63

C. Transmittance Properties of Simulated Bimodal Coatings

In dilute systems the particles of two sizes act as independent scatterers. It was relatively easy to obtain the experimental evidence for the dilute suspensions, since the physical parameters of the scattering system (such as particle size, concentration or separation of scatterers, optical path length) were relatively well defined. As the first approximation to a real pigmented paint, a series of concentrated mixtures containing two particle sizes of silver bromide in gelatin were prepared. A complete description of the silver bromide batches (particle diameter, size distribution, precipitation conditions) was given in our last report.⁹ These bimodal mixtures were deposited as thin films on quartz plates. The spectral transmittances of the mixtures were compared with the transmittances of similar films prepared from single particle sizes. In each case, a set of three films was compared, two obtained from monodisperse suspensions and one from the mixture.

The optical densities obtained from spectral transmittance measurements for the two monodisperse films are

⁹IIT Research Institute, loc. cit.

$$D_1 = K\pi \left[(r_1)^2 n_1 l_1 \right] \quad (1)$$

and

$$D_2 = K\pi \left[(r_2)^2 n_2 l_2 \right] \quad (2)$$

where K is the total Mie scattering coefficient, r is the particle radius, n is the particle concentration, l is the optical path length or the thickness of the films, and D is the optical density.

If the two particle sizes act as independent scatterers, the transmittance of a hypothetical sum of two monodisperse films at 50% of the original concentration is

$$\frac{D_1 + D_2}{2} = K\pi \left[(r_1)^2 \left(\frac{n_1 l_1}{2} \right) + (r_2)^2 \left(\frac{n_2 l_2}{2} \right) \right] \quad (3)$$

If it is again assumed that the particles act as independent scatterers in the mixed suspension, the optical density of the mixture (at 50/50% by weight) is

$$D_{1+2} = K\pi \left[(r_1)^2 \left(\frac{n_1}{2} \right) + (r_2)^2 \left(\frac{n_2}{2} \right) \right] l_{1+2} \quad (4)$$

where l_{1+2} is the thickness of the film prepared from a mixed suspension. If

$$l_1 \approx l_2 \approx l_{1+2}$$

then

$$\frac{D_1 + D_2}{2} \approx D_{1+2}$$

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This method was used to construct a series of curves predicting the optical properties of bimodal mixtures from the single particle observations. These curves were compared with those obtained experimentally. The data are summarized in Figures 3 to 8, indicating that the two size particles act as independent scatterers for very thin concentrated films.

It should be emphasized that the above observation is valid probably for not more than several layers of particles. Also, the greatest source of error in our experimental measurements is the determination of thickness and concentration of scatterers in dry films. The thickness was measured interferometrically as described in our last report.¹⁰ An appreciable displacement of the observed and theoretically constructed curves due to variations in thickness and concentration can be expected. The thickness measurements are summarized in Table 2.

Table 2
THICKNESS OF FILMS USED IN EXPERIMENTAL STUDIES

<u>Films</u>	<u>Batch No.</u>	<u>Particle Radius, r</u>
Monodispersed	25	0.83 - 1.00
	26	0.54 - 0.60
	28	0.72 - 1.20
	30	0.66 - 0.77
	31	0.54 - 0.66
Mixtures	25, 30	0.66 - 0.96
	28, 30	0.60 - 0.66
	26, 30	0.60 - 0.78

¹⁰IIT Research Institute, loc. cit., p. 28.

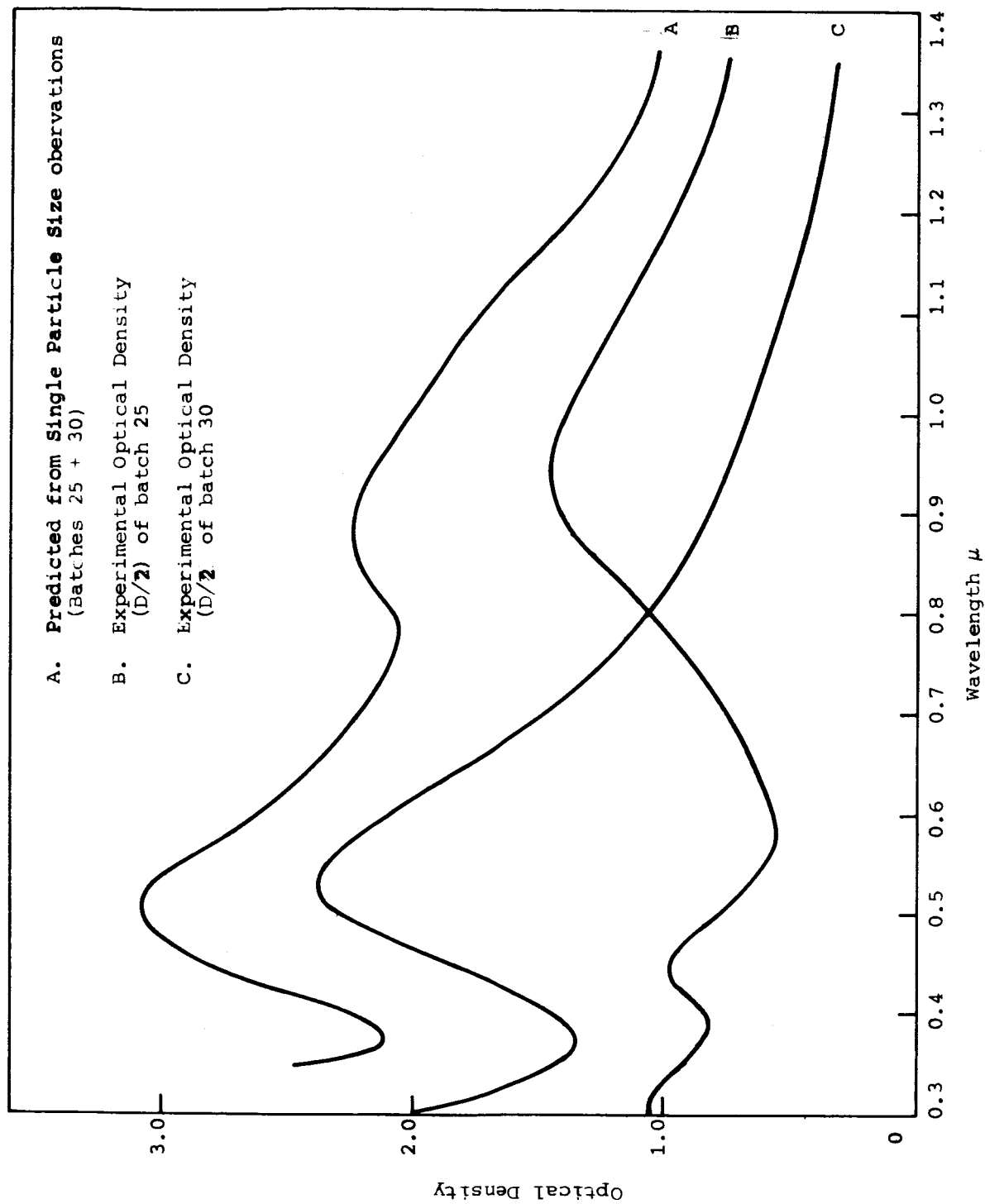


Figure 3

OPTICAL DENSITY OF SILVER BROMIDE SUSPENSIONS (BATCHES 25 AND 30)

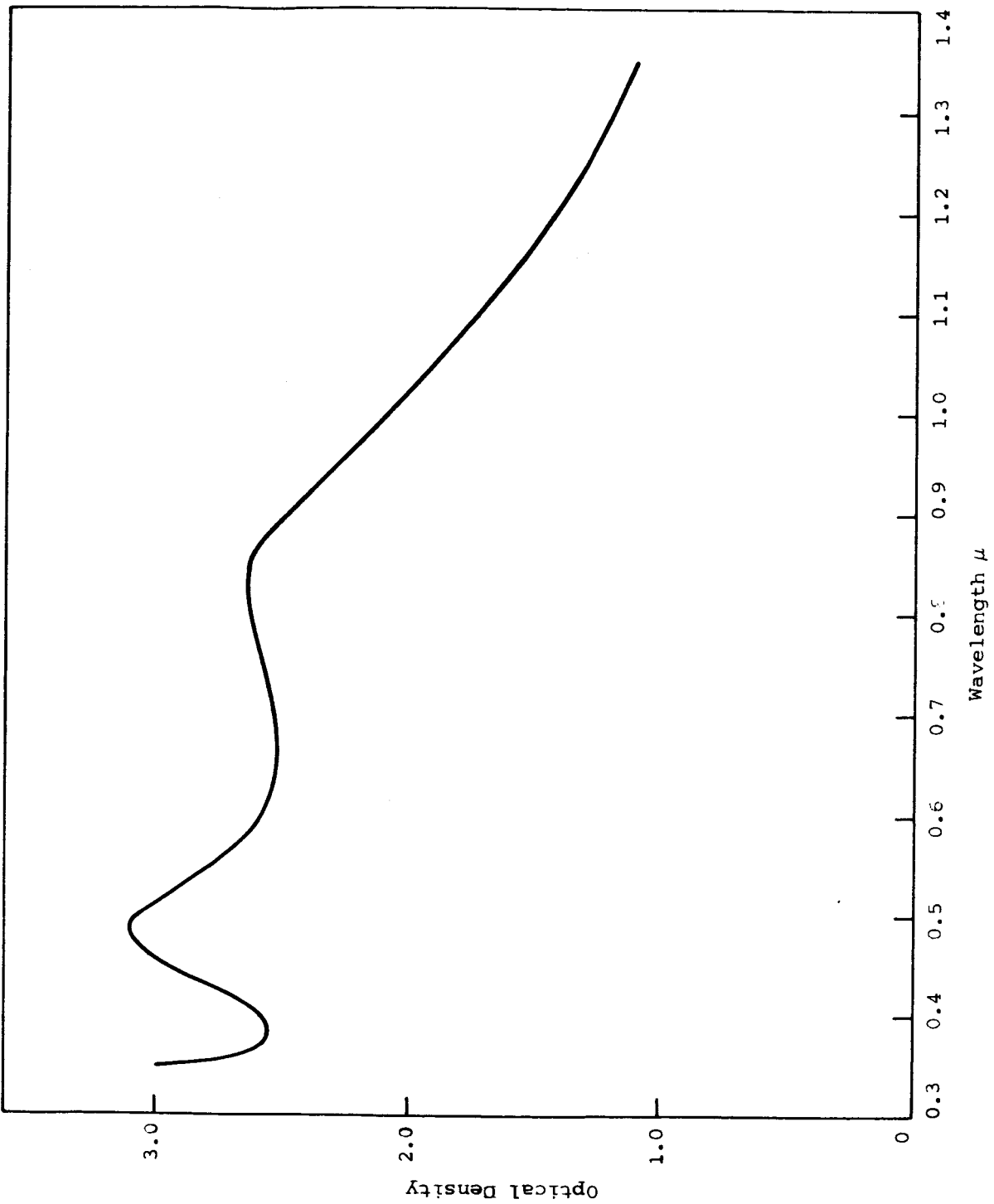


Figure 4

EXPERIMENTALLY OBSERVED OPTICAL DENSITY OF A 50/50 MIXTURE OF BATCHES 25 AND 30

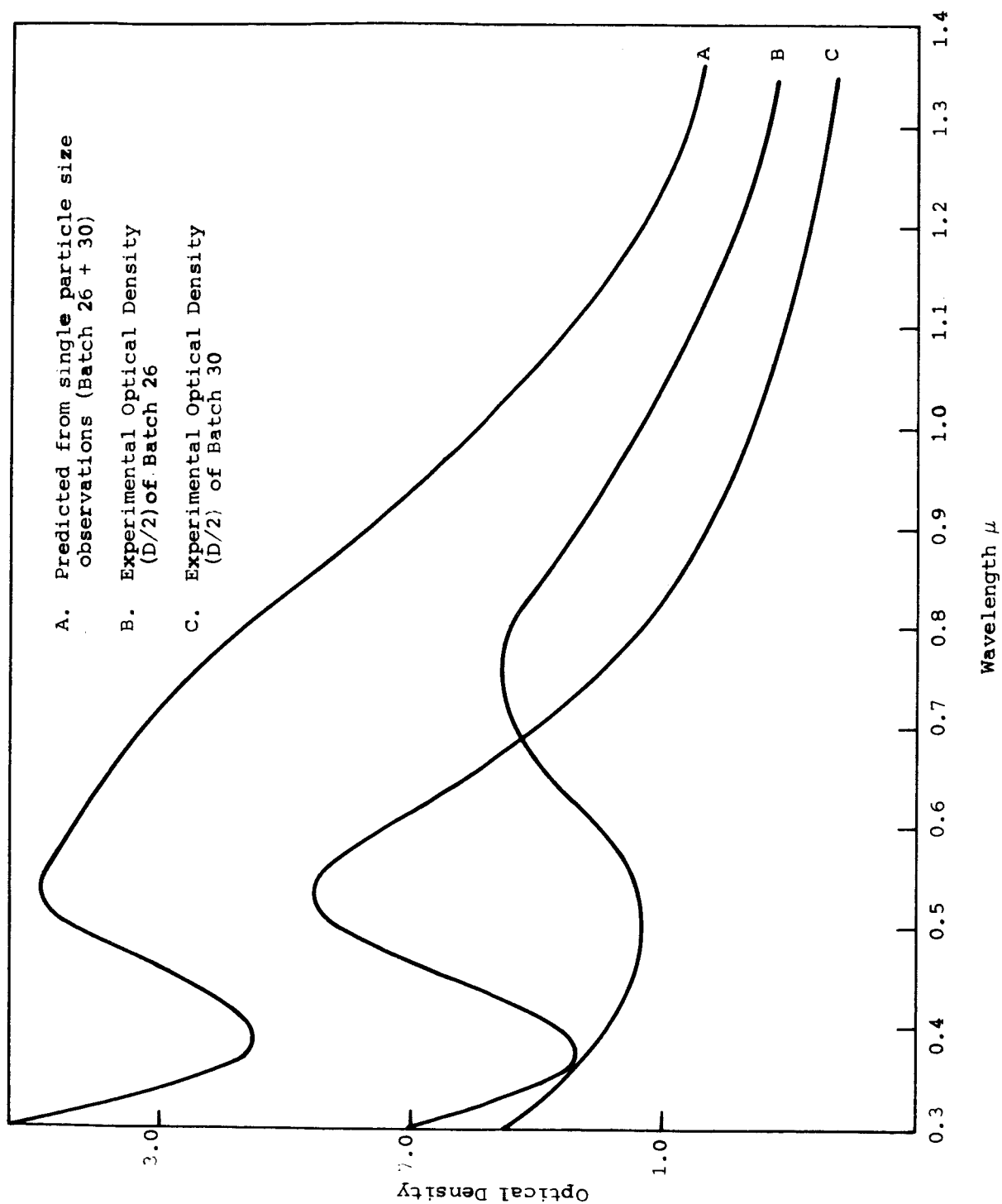


Figure 5

OPTICAL DENSITY OF SILVER BROMIDE SUSPENSIONS (BATCHES 26 AND 30)

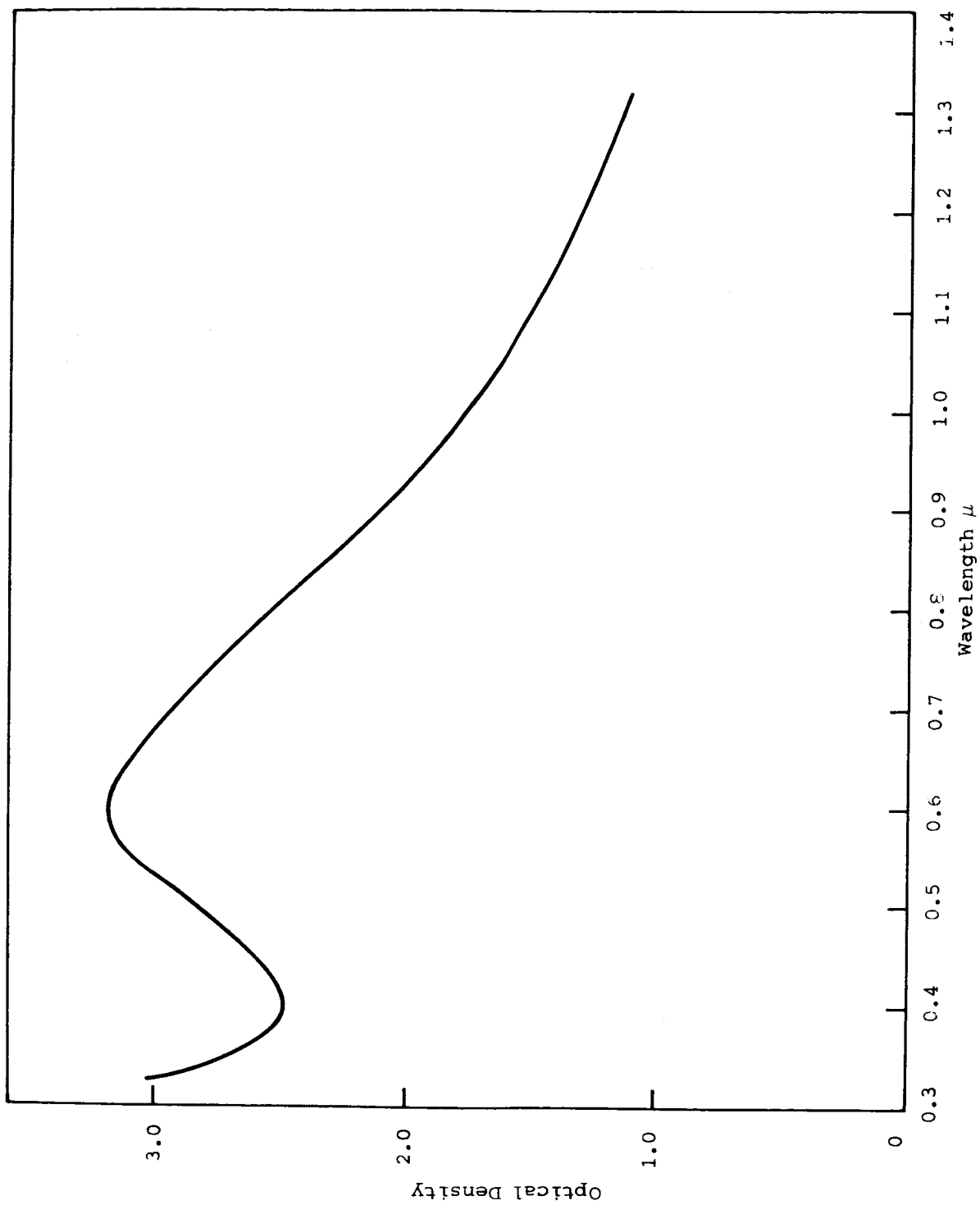


Figure 6

EXPERIMENTALLY OBSERVED OPTICAL DENSITY OF A 50/50 MIXTURE OF BATCHES 26 AND 30

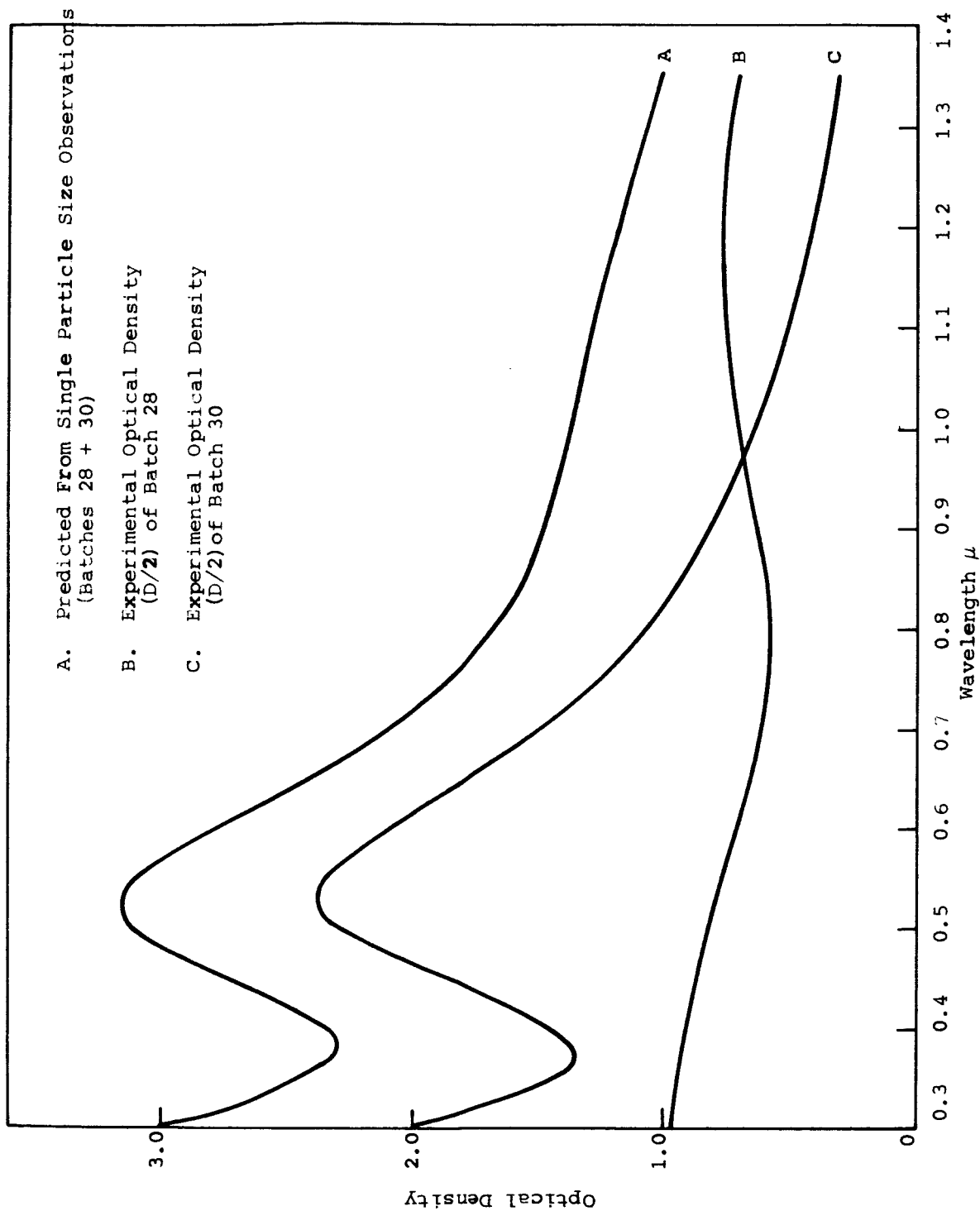


Figure 7

OPTICAL DENSITY OF SILVER BROMIDE SUSPENSIONS (BATCHES 28 AND 30)

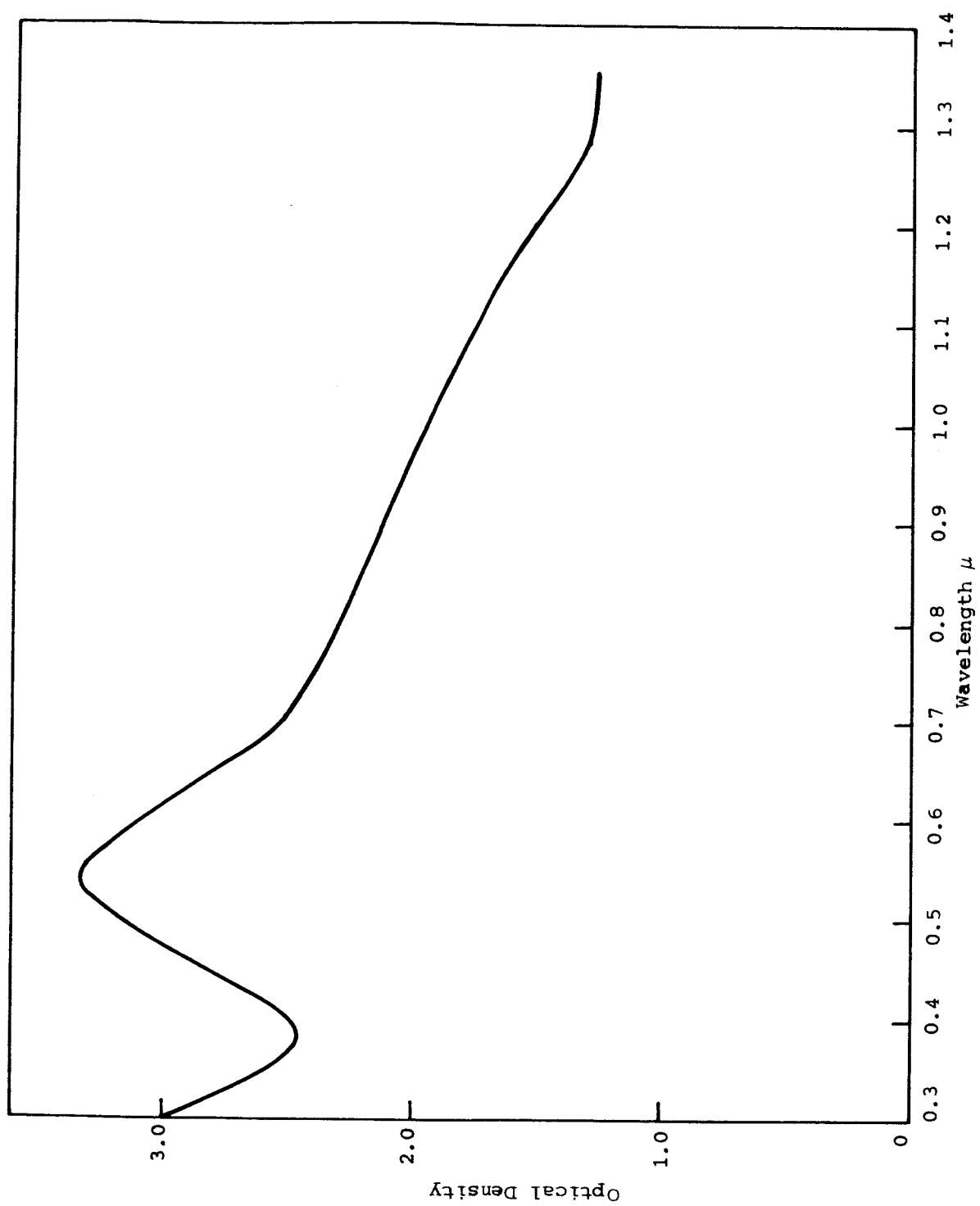


Figure 8

EXPERIMENTALLY OBSERVED OPTICAL DENSITY OF A 50/50 MIXTURE OF BATCHES 28 AND 30

D. Transmittance Properties of a Theoretical (Mie) Bimodal Coating

Since a reasonable agreement was obtained between the observed transmittances of the bimodal mixtures and those constructed from observations of single particles, the above comparison was extended to include a theoretical bimodal coating constructed on the basis of Mie scattering theory. An inherent assumption in such a case is that the particles are completely uniform -- there is no size distribution whatsoever for any given particle size.

Theoretical variation in minima and maxima with the width of the distribution curve is given in Figure 9 which was reported by Stevenson, Heller, and Wallach.¹¹

A comparison of observed optical densities and graphically constructed theoretical ones for several particle sizes is given in Figures 10 and 11.

The comparison of observed and theoretical bimodal coatings is given in Figures 13 and 14. The data indicate that although there is some loss of fine structure, the agreement between observed and calculated curves is quite good. The loss of several lobes, which was predicted by the theory, is obviously due to the width of the distribution curves of the particles used in the bimodal mixture.

¹¹Stevenson, A. F., Heller, W., and Wallach, M. L., Journal of Chemical Physics, Vol. 34, p. 1789, 1961.

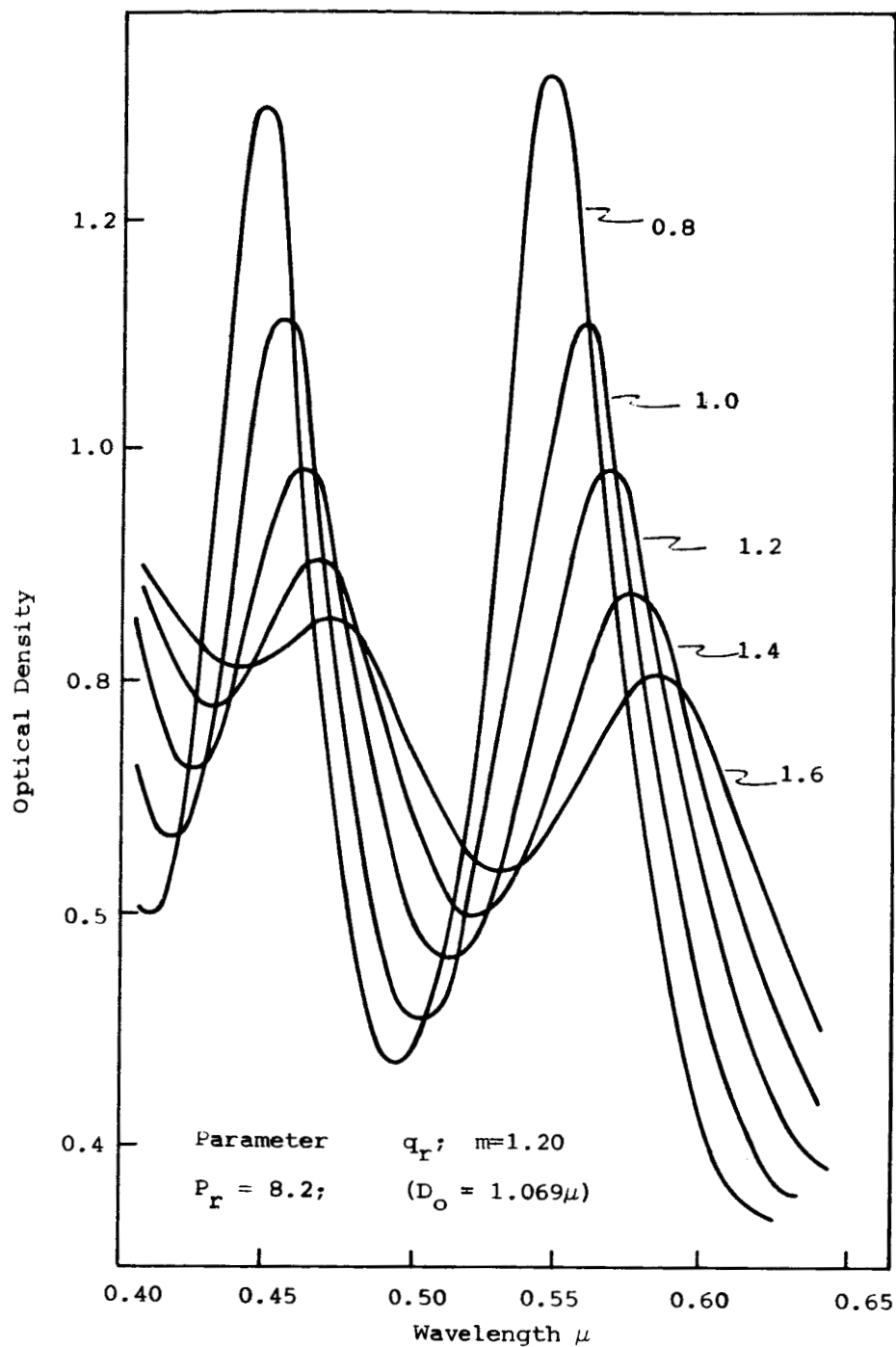


Figure 9

THEORETICAL VARIATION OF MAXIMA AND MINIMA OF SCATTERING RATIO WITH INCREASE IN WIDTH OF THE SIZE DISTRIBUTION CURVE AS EXPRESSED BY INCREASING q_R -VALUES: INCREASE IN q_R MEANS AN INCREASE IN THE WIDTH OF THE DISTRIBUTION CURVE

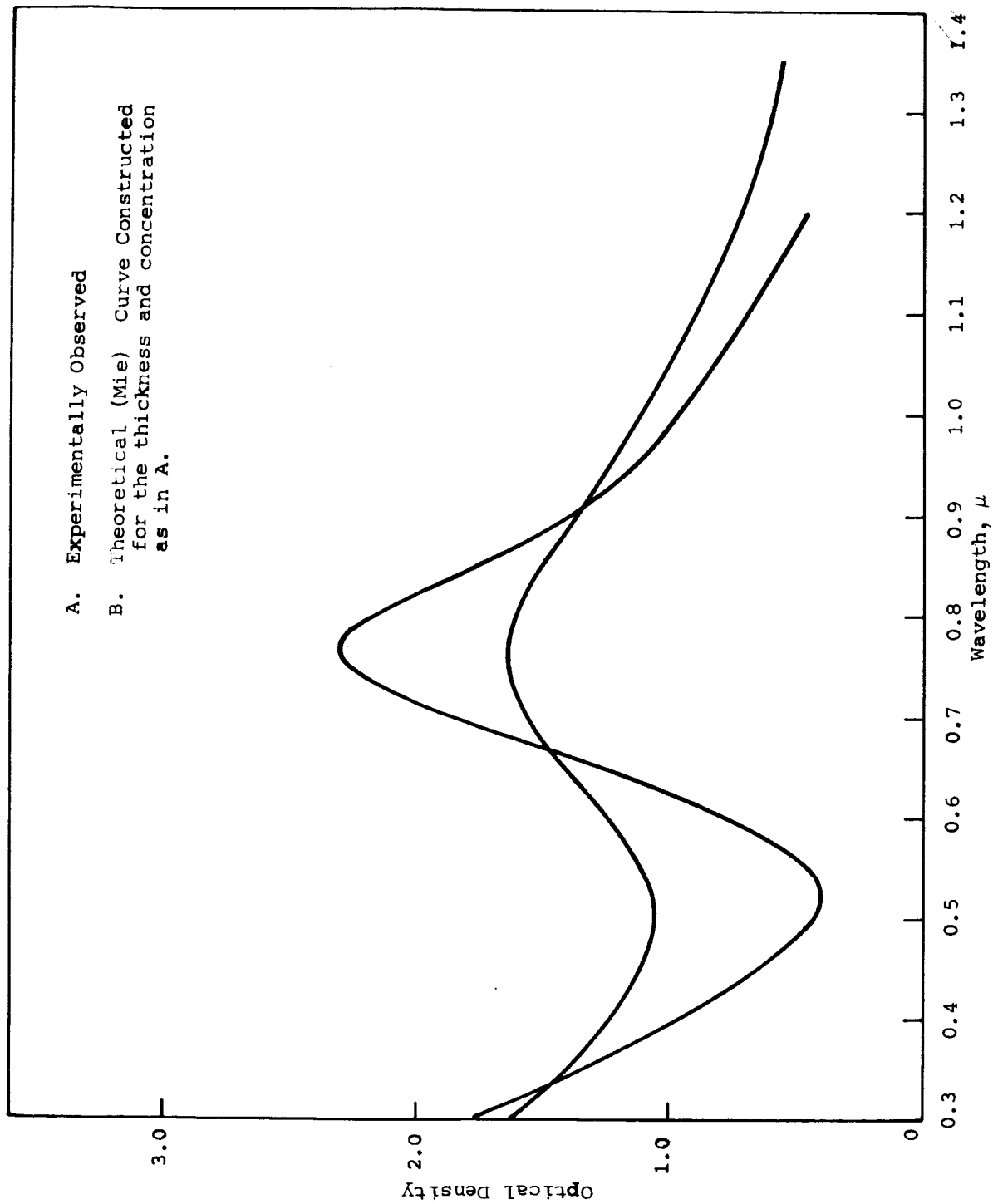


Figure 10

OPTICAL DENSITY OF MONODISPERSE FILM OF BATCH 26

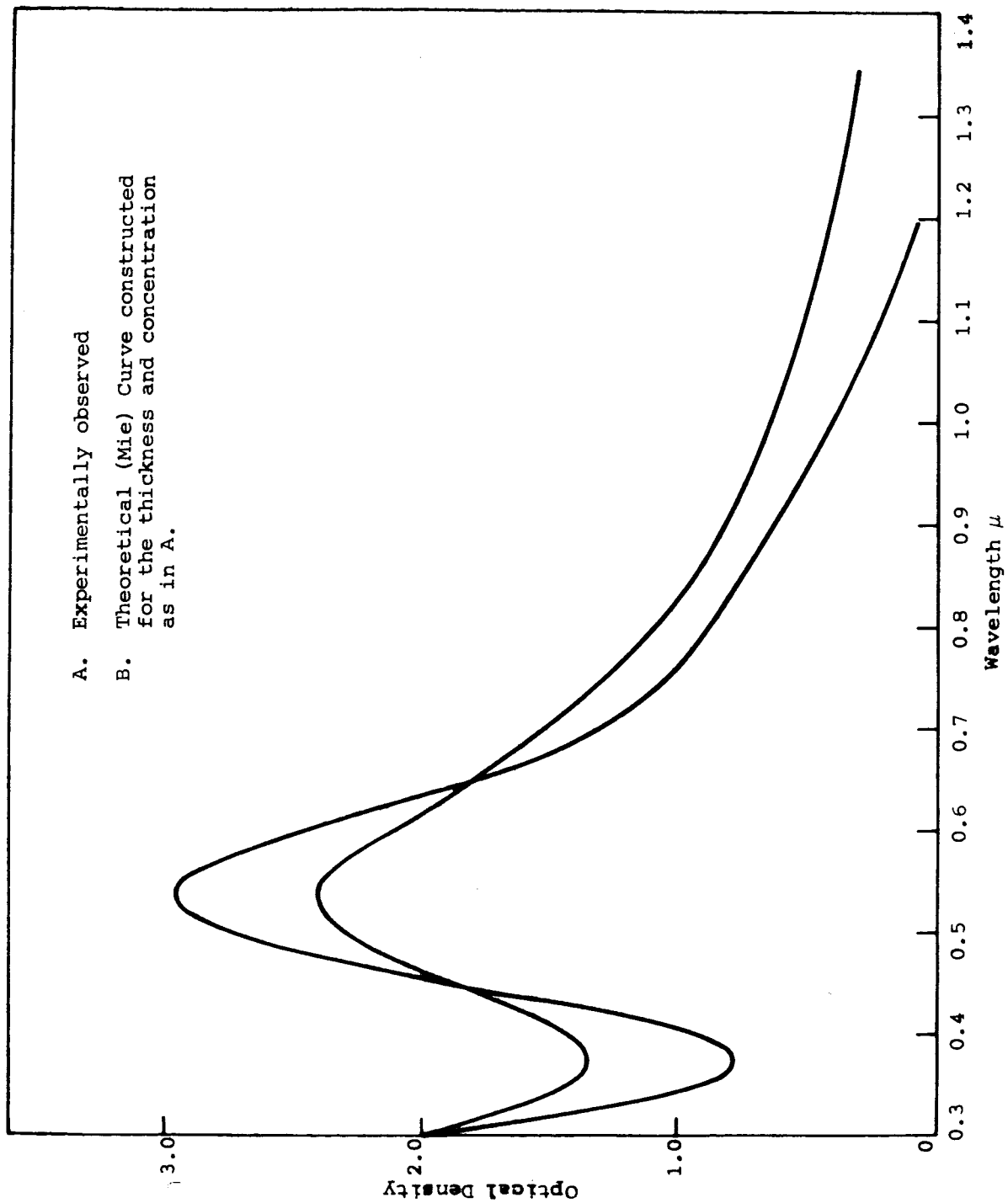


Figure 11

OPTICAL DENSITY OF MONODISPERSE FILM OF BATCH 30

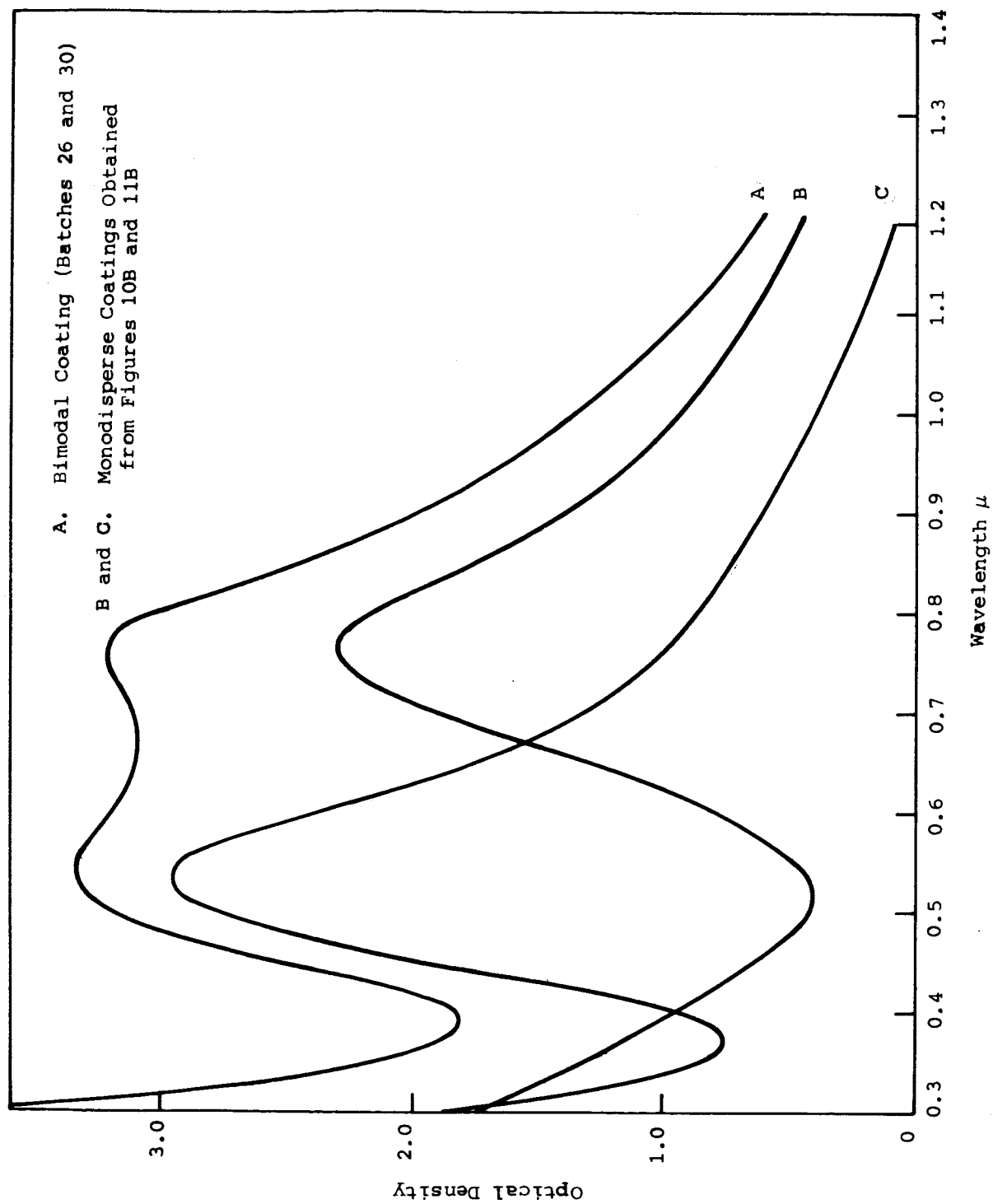


Figure 12

THEORETICAL (MIE) DENSITY OF SILVER BROMIDE COATINGS (BATCHES 26 AND 30)

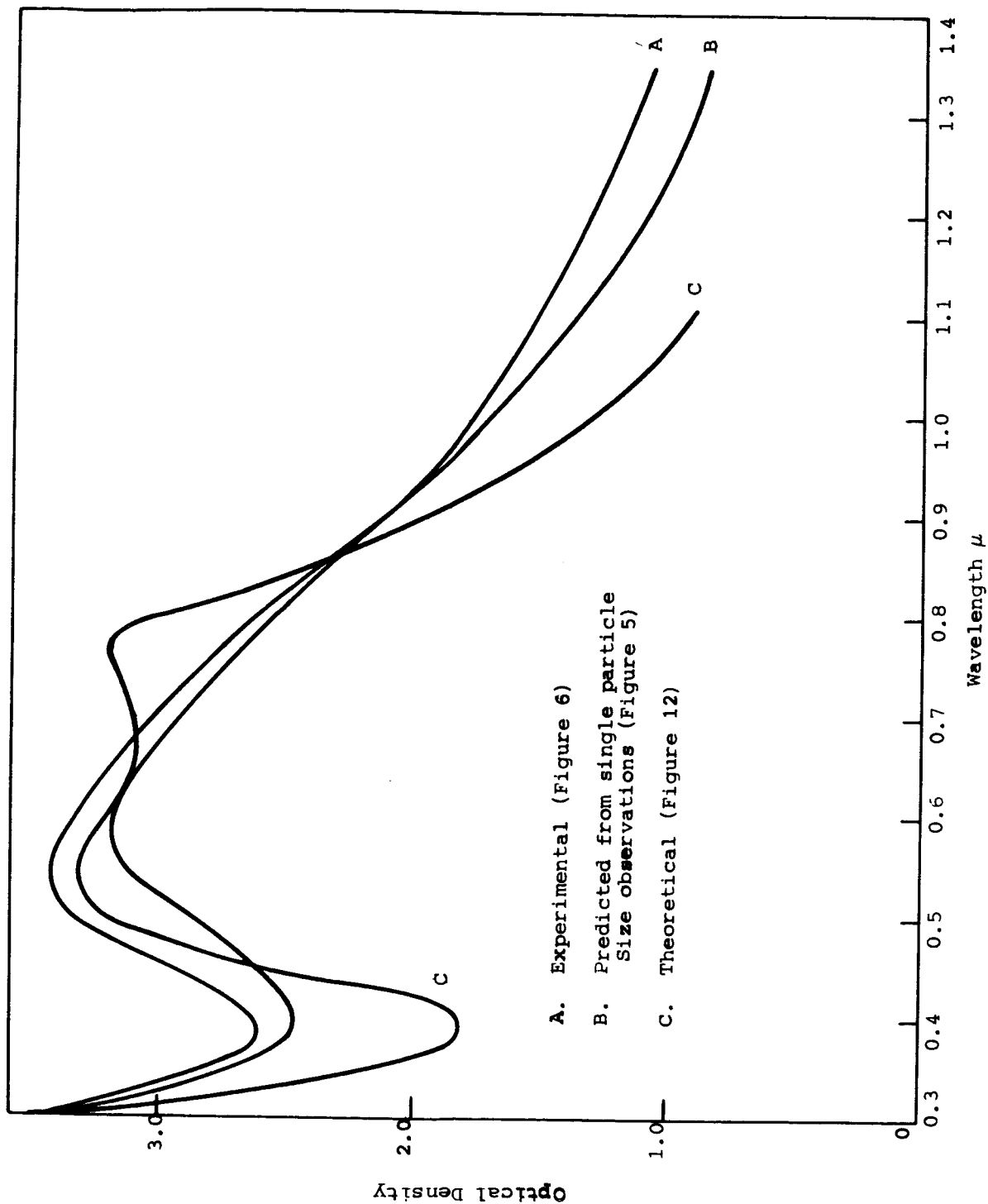


Figure 13

OPTICAL DENSITY OF A MIXTURE OF BATCHES 26 AND 30

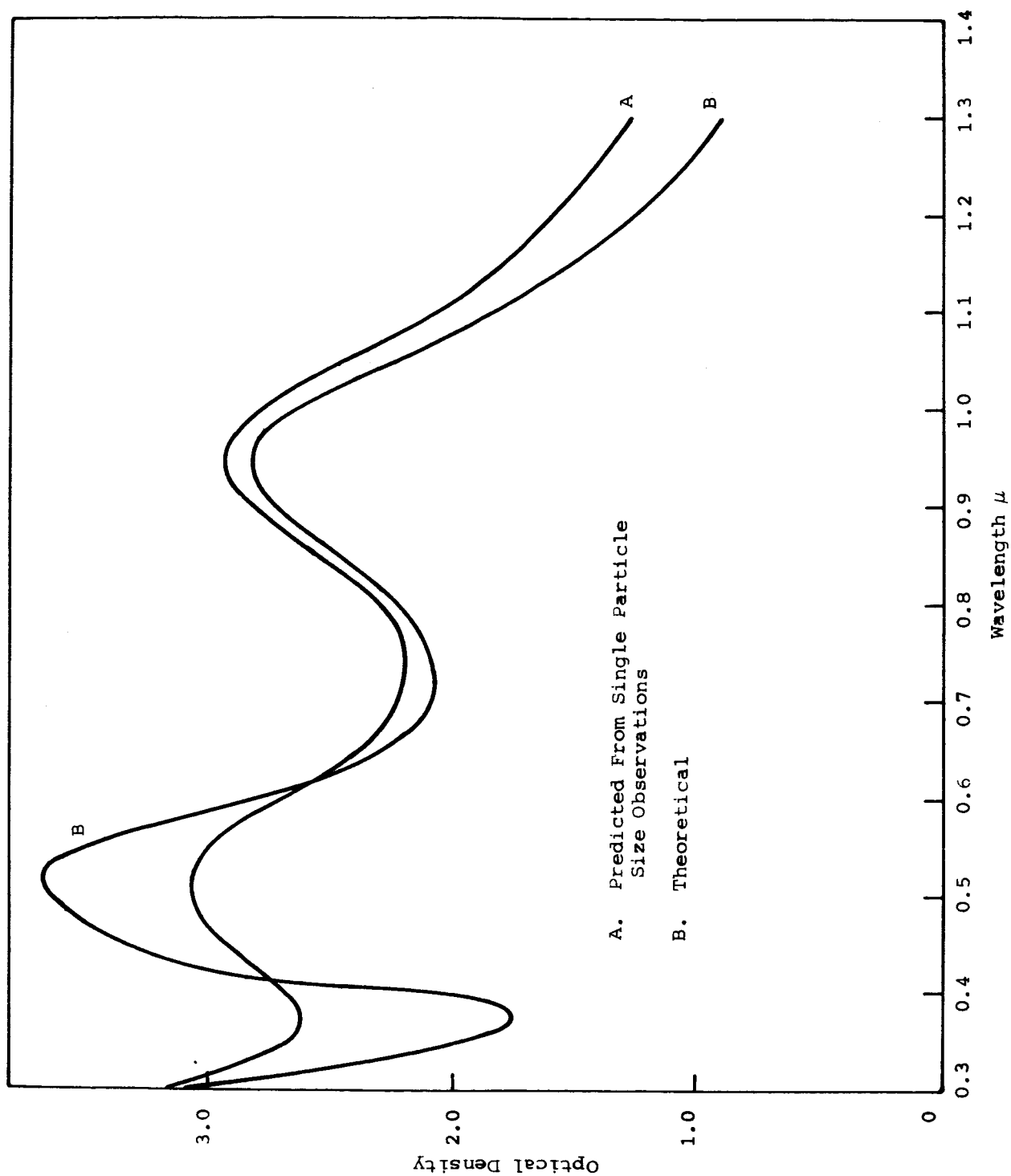


Figure 14

OPTICAL DENSITY OF A MIXTURE OF BATCHES 25 AND 30

E. Discussion

Previous experimental results indicate that a theoretical model based on Mie scattering theory or a semi-empirical model based on single particle observation will predict the spectral transmittance properties of thin pigmented films to a fairly close approximation. The effects of absorption and interface reflection have not yet been considered directly in the experimental studies. It appears that the absorption effects are closely related to multiple scatter, as noted in our previous report.¹² The effects of multiple scatter and absorption become quite significant in films of several layers of scatterers (i.e., more than 5). Thus, in any real pigmented paint system some degree of multiple interactions and absorption will always be present, but these effects are minimized in thin layers of monodisperse suspensions.

Thus, based on our experimental data (which so far consist primarily of spectral transmittance measurements) a real pigmented coating should fulfill the following set of requirements

1. The substance to be used as scatterer (pigment) should have a minimum bulk absorption coefficient for any given spectral band. Less radiant energy will inherently be lost and converted to thermal and molecular energy modes, if the scattering particles are transmitting.
2. The ratio of refractive indices between particles and suspension media should be the largest available. Total scattering and especially back-scattering

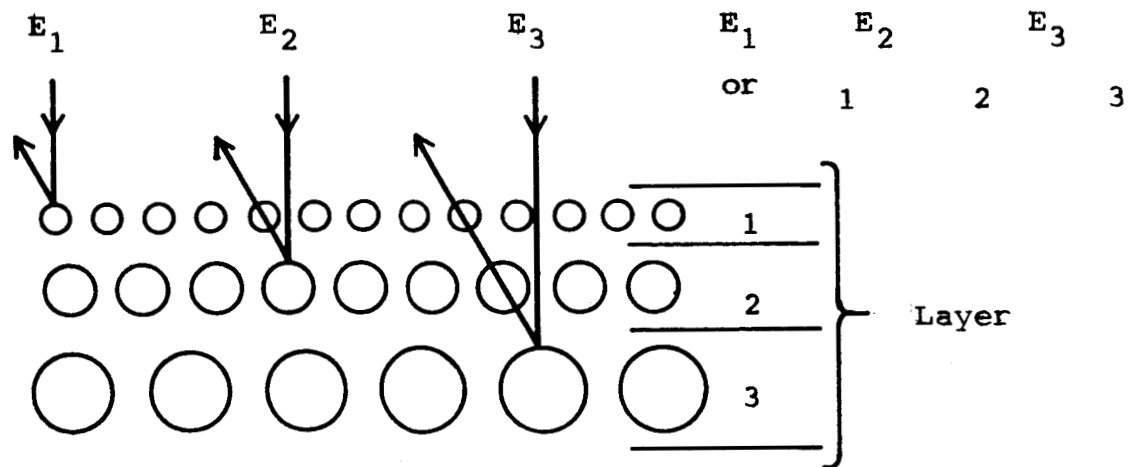
¹²IIT Research Institute, loc. cit., p. 31.

efficiencies increase very significantly at high effective refractive indices, with the exception of the refractive index which is close to infinity. In this case, the radiation can no longer penetrate the particle, and this lack of optical resonance greatly reduces the scattering efficiency.

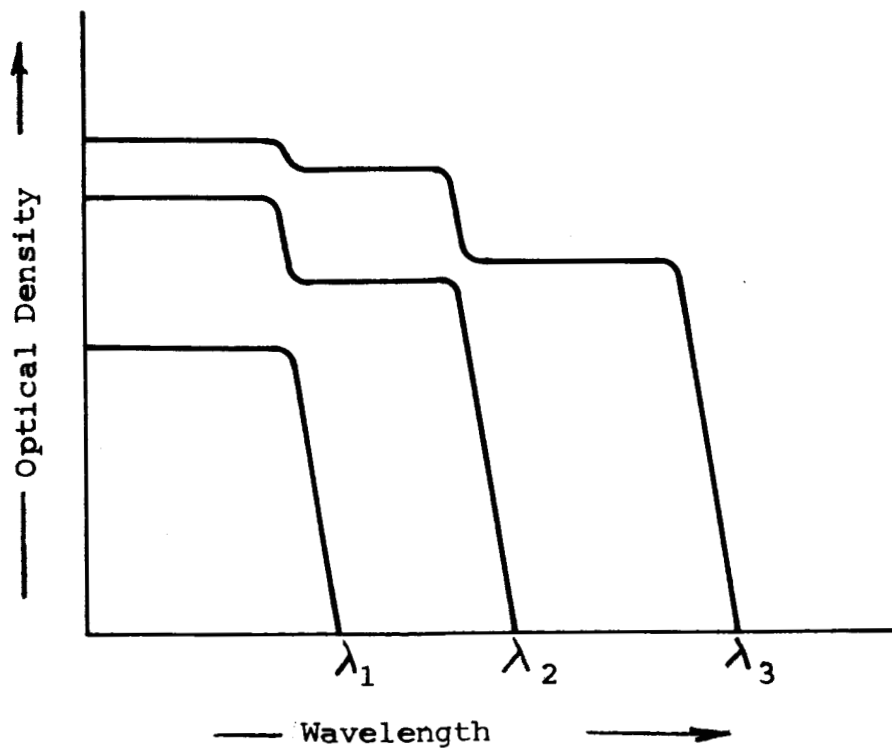
3. To attain minimum interactions between the scatterers, the particles should be packed in exceedingly thin layers containing particles of uniform size. The distribution of layers with respect to the incident beam should be such that each layer contains increasingly larger particles. The top layer, containing the smallest particles, will interact only with shortest wavelengths, allowing longer wavelengths to pass undisturbed in both directions. In this manner, each succeeding layer will interact with radiation of increasing wavelength. Such a coating is schematically illustrated in Figure 15.

F. Validity of Transmittance Measurements

In the measurements of the transmittance of a scattering system, neglecting absorption and interface reflection, the total energy loss due to redirection of photons is measured. However, in a real reflecting paint surface the vital interest is the amount of energy that is back-scattered with respect to the primary beam of radiation. In a reflecting coating, therefore, the back reflectance is probably best expressed in terms of integrated reflectance measurements. Our next step should be a formulation of a scattering model of a coating based on the integrated reflectance measurements of monodisperse films. It should be pointed out that the requirements for an optimum coating stated previously (which are based on the transmittance



Interactions between Protons
and Layers at Increasing Particle
size



Optical Density Due to Successive
Addition of Layers

Figure 15

SCHEMATIC OF MULTILAYER COATING AND
RESULTANT OPTICAL DENSITY OF LAYERS

measurements) should be valid for the reflectance or back-scattering model because, in general, back-scatter depends on the total amount of scattering.

In our previous reports, we stated the desirability of investigating the radial distribution of scattered light. It has been found that such an investigation (of dilute silver bromide suspensions) was carried out by Napper and Ottewill¹³ and others. Some of the data is given in Figures 16 to 18. The data confirms our observation that in dilute suspensions the distribution of intensities predicted by Mie theory is quite closely approximated in experiments. During the next research period, our immediate experimental objective will be the establishment of relationships between physical and optical parameters from the measurements of the integrated reflectance of monodisperse and bimodal coatings.

¹³Napper, D. H. and Ottewill, R. H., "Electromagnetic Scattering," The MacMillan Company, New York, p. 377, 1963.

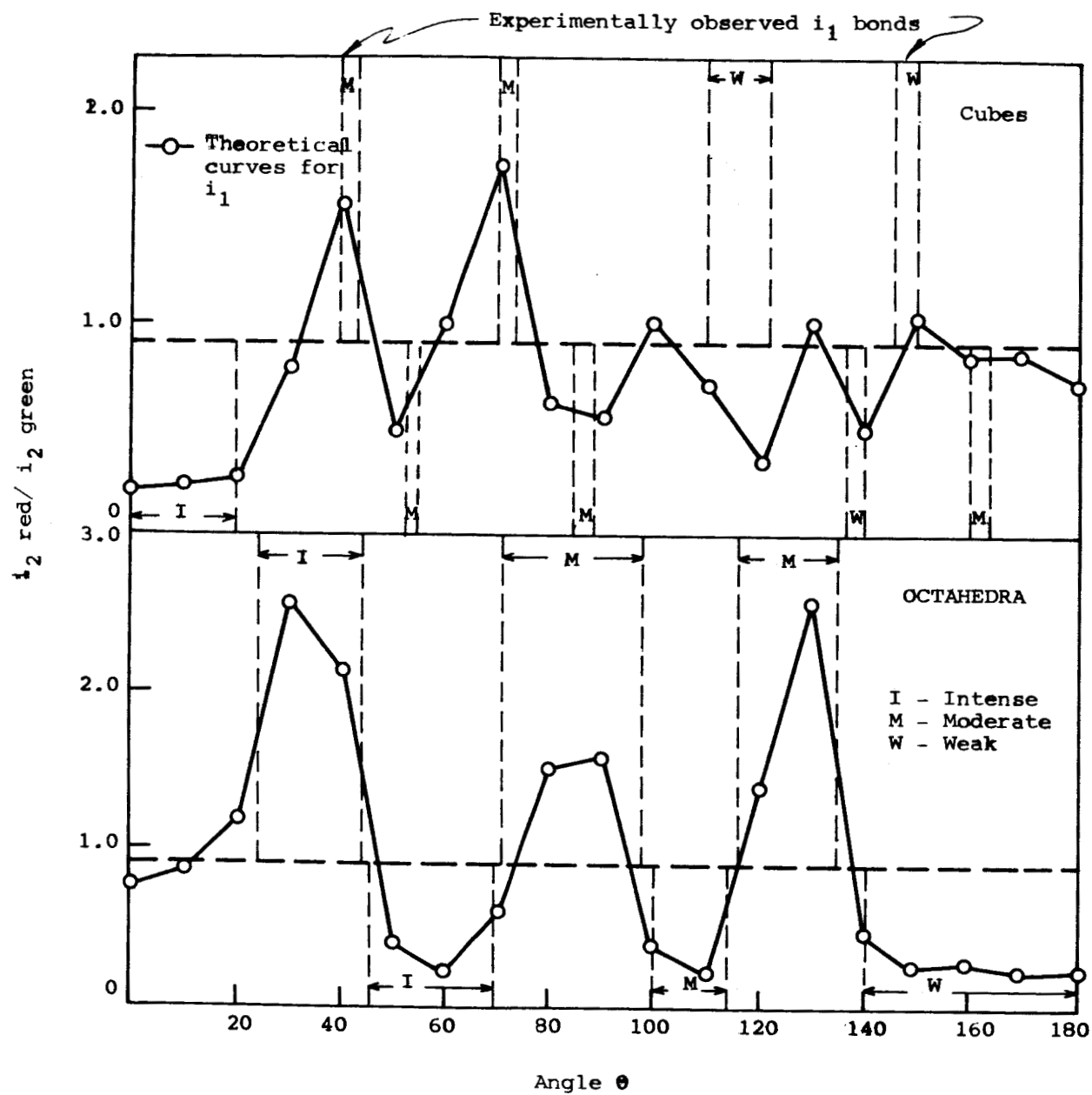


Figure 16
HIGHER ORDER TYNDALL SPECTRA

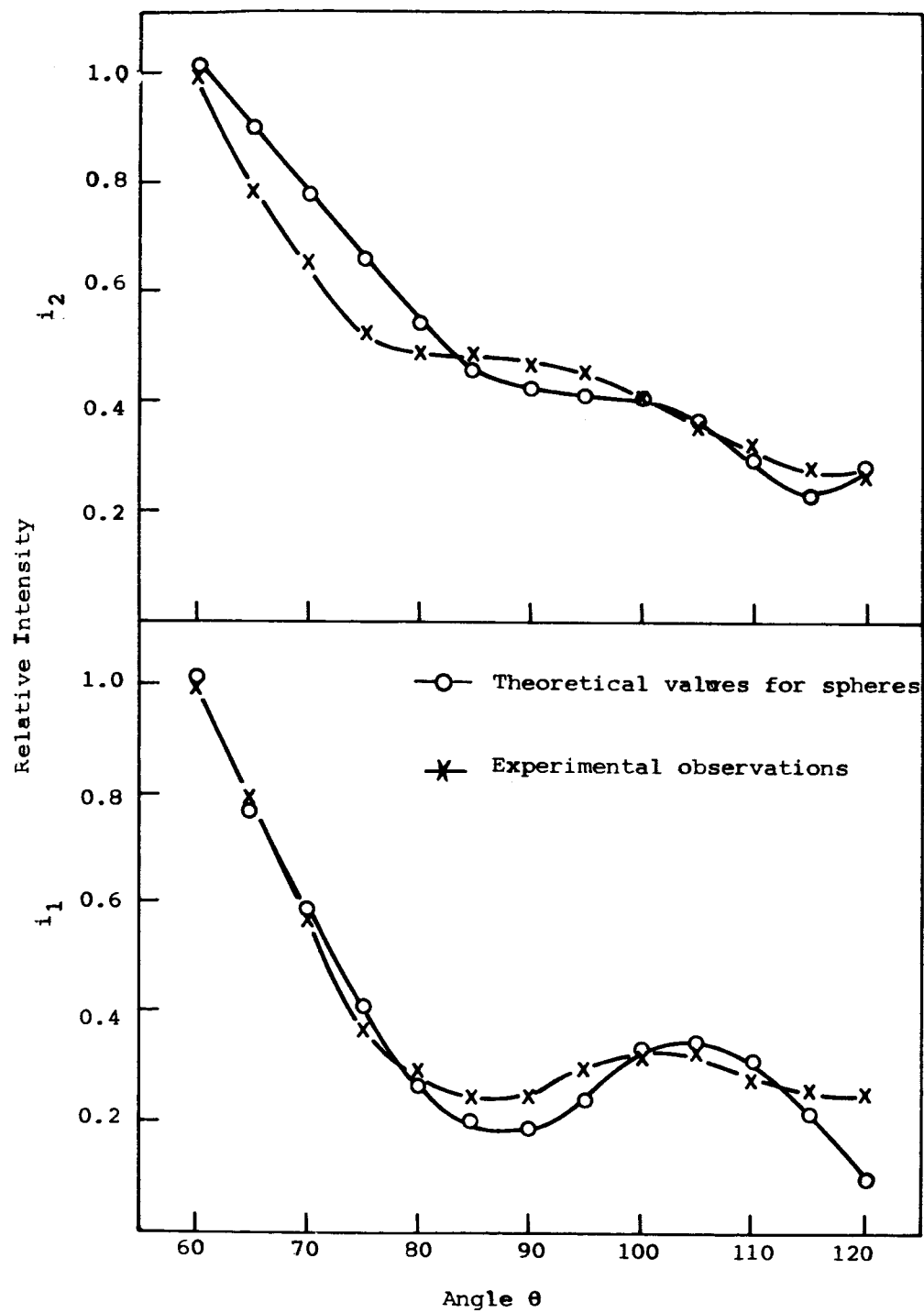


Figure 17

RELATIVE INTENSITY OF SCATTER VERSUS ANGLE FOR SOLS OF OCTAHEDRAL SILVER BROMIDE

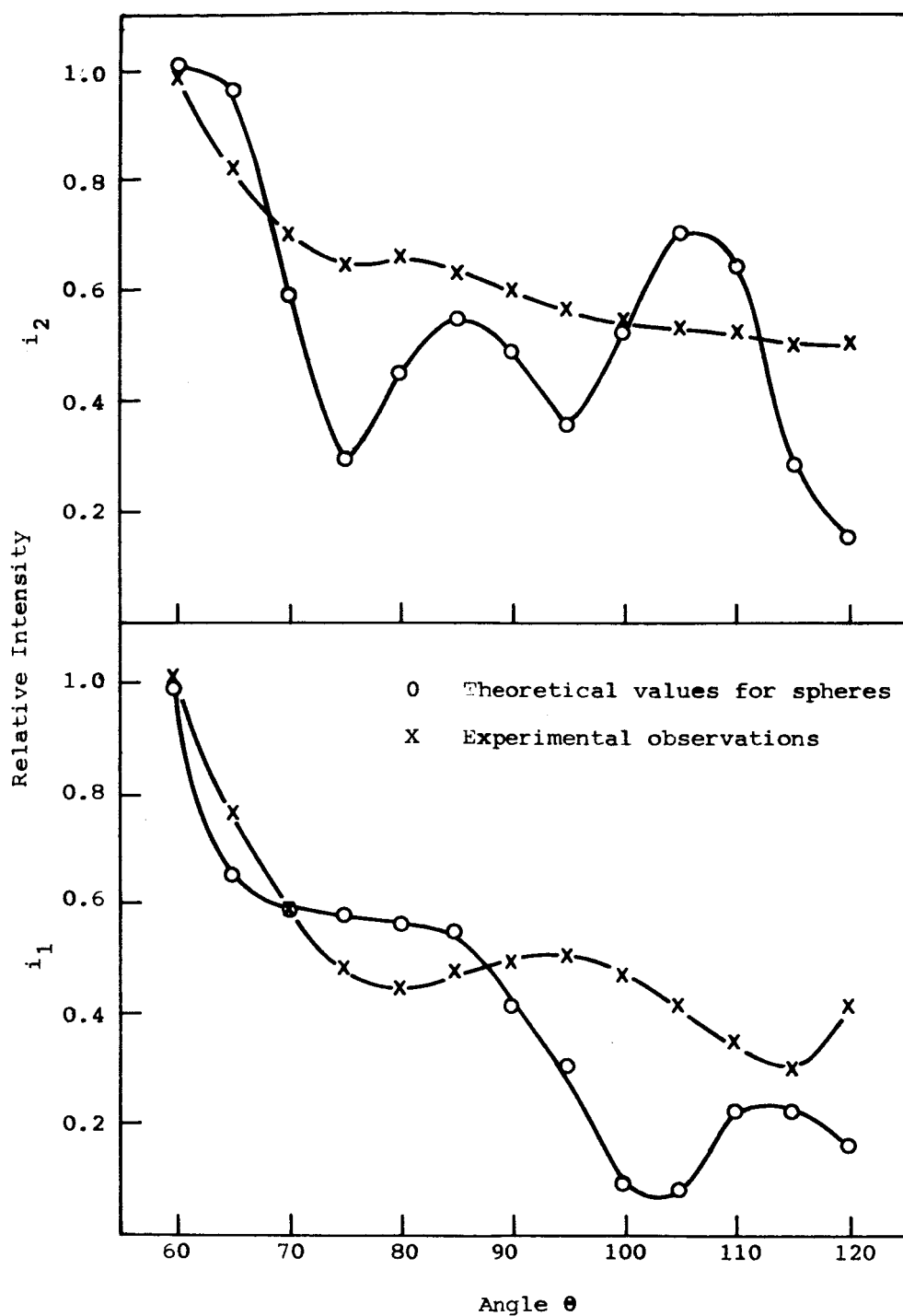


Figure 18

RELATIVE INTENSITY OF SCATTER VERSUS ANGLE FOR SOLS OF CUBIC SILVER BROMIDE

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